THE DETERMINATION OF HEAVY METALS IN WATER FLOWED WITH METAL WASHING WASTE

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ABSTRACT

The water stream in Desa Kejapanan, Pasuruan, East Java, Indonesia used as a source of water for households and field irrigation is indicated to experience heavy metal contamination. The purpose of this study is to determine the type and concentration of metals in the water stream that is contaminated with metal washing waste. The concentration of heavy metals in water and sediments was tested using the Atomic Absorption Spectrophotometer (AAS) method. In this case, the physical and chemical components of the water measured were color, temperature, pH and DO. Meanwhile, the data of heavy metal concentration was tested using ANOVA and was analyzed descriptively. The data were analyzed using SPSS version 16.0 software. The test results point out that the concentration of mercury (Hg) is ranged from 0.028±0.002 mg/L to 0.036±0.005 mg/L while lead (Pb) is in between 0.492±0.009 mg/L and 0.665±0.007 mg/L, cadmium (Cd) in between 0.142±0.011 mg/L and 0.161±0.004 mg/ L, zinc (Zn) in between 0.737±0.006 mg/L and 0.763±0.004 mg/L), and copper (Cu) in between 0.318±0.004 mg/L and 0.369±0.002 mg/L. It is known that Zn has the highest concentration of metals compared to other metals while the highest concentration of all types of metals (Hg, Pb, Cd, Zn, and Cu) occurs in the sample S2. The water stream is indicated to have been contaminated by heavy metals (Hg, Pb, Cd, Zn, Cu) because the concentrations of these metals have exceeded the maximum gualification both in water and sediments.

KEY WORDS

Waste, heavy metals, polluted waters.

Aquatic ecosystems are an open ecosystem that is possible to be polluted by heavy metals (Mohiuddin, Ogawa, Zakir, Otomo, & Shikazono, 2011). The heavy metals in the aquatic system come from natural and anthropogenic sources (Lohani et al., 2008) that are generated from domestic and industrial waste disposal (Duruibe, Ogwuegbu, & Egwurugwu, 2007 : Nagaivoti et al., 2010) as well as agricultural waste (Risiani et al., 2014). The heavy metals increases due to the entry of wastes from industries and other activities (Liu et al., 2006). In fact, these wastes are found in sediment and water. The sources of elements that pollute the water can be found through the weathering of sedimentary rocks such as limestone, dolomite, shale, and sandstone. The interaction of water with igneous rocks contributes to several main elements such as Fe, Cu, Al, Zn, Mn (Ball and Izbick 2004, Viers et al., 2007, Borrok et al., 2004, Huffmeyer et al., 2009). The elements above are concentrated in mineral deposits which contain sulfides especially those which are related to gold mineralization and iron ore (Nordstrom, 2002). Some minor elements such as Cd, Co, Mn occur in the earth's crust with other minerals (Jarup, 2014). Pb and Hg are sedimented into the aquatic system of atmospheric aerosols that is formed from dust, emissions volcanic, forest, and plant fires (Cempel and Nickel, 2006, Hsu et al., 2010, Kang et al., 2011, Li, 2014).

Heavy metals are harmful substances because it has toxic nature (Duruibe et al., 2007), nondegradable (Ahluwalia & Goyal, 2007), and bioaccumulative (Shi, M, Zhang, Jiang, & Li, 2010). Since heavy metals cannot be degraded, precipitation will occur in water, sediments, and aquatic organisms (Nagajyoti et al., 2010). Heavy metals enter the water stream naturally and become part of the water and sediment suspension through the process of absorption, precipitation, and ion exchange (Liu et al., 2010). The concentration of heavy metals in sediments is much higher than in water. In general, there is a process of sedimentation in river estuary where heavy metals that are hard to dissolve undergo a dilution process in water, drop to the bottom of the water stream, and settle in the sediment (Facetti et al., 1998). Heavy metals in water by natural sources depend on local geology, hydrogeology, and geochemical characteristics of aquifers (Wang et al., 2006).

In Indonesia, the problem of heavy metal pollution is not only generated from the industrial sector but also from agricultural and households sector (Murtini et al., 2003). In general, the heavy metals which can be found in polluted water are mercury (Hg) (Soegianto, 2007), lead (Pb) (Koedrith and Seo, 2011), cadmium (Cd) (De Recherche and Es, 2013), chromium (Cr) (Rodríguez et al., 2016), and copper (Cu) (Asztalos, Italiano, Milano, Maróti, & Trotta, 2010). It is reported that mercury, lead, and cadmium are heavy metals that highly pollute the water in Indonesia (Siregar & Murtini, 2008) that one of which can be seen in Sungai Wangi, Pasuruan (Adam, Maftuch, Kilawati, Tahirah, & Risjani, 2018).

This research is concentrated on analyzing the water in Desa Kejapanan, Pasuruan, East Java that is used as a source of water for household activities and paddy field irrigation. However, the water has flowed with metal washing waste, households waste, and industrial waste that is indicated to be contaminated with heavy metals. Heavy metals such as mercury (Hg), cadmium (Cd), chromium (Cr), lead (Pb), copper (Cu) have high toxicity and will accumulate in the body of the organism (Murty et al. 2011). Because of the important role of water, studies that are related to the content of heavy metal in water are needed. Therefore, the purpose of this study is to determine the type and concentration of metal in the water that is polluted with metal washing waste.

MATERIALS AND METHODS OF RESEARCH

There were water and sediment samples taken from four sampling points (S1, S2, S3, and S4) where S1 sampling point was located before the pollutant source and S2 was right at the source of pollution. On the other hand, S3 and S4 sampling points were located after the pollutant source. Water samples were collected by using a BOD Bottle (250 ml) whereas sediment samples were taken using PVC. Then, the samples were inserted into the bottom of the water as deep as 20 cm and stored in containers (Jamabo & Chinda, 2010) until the metal test took place.

Mercury (Hg), cadmium (Cd), lead (Pb), zinc (Zn), and copper (Cu) concentrations in water and sediments were tested by using the *Atomic Absorption Spectrophotometer* (AAS) method. In this study, the procedure of metal concentration determination refers to EPA (2007). Up to 50 mL of the samples were taken and 5 mL of concentrated HNO₃ was added. Then, it was heated slowly until the volume remained 15-20 mL. After that, 5 mL of HNO₃ was added and heated again followed by adding acid and heated it one more time until the mercury dissolved. The test samples were taken for about 10 mL and then added with KmnO₄ 0.01N and put into a spectrophotometer with a wavelength of 253.6 nm (Hg), 228.8 nm (Cd), 283.3 nm (Pb), 213.9 nm (Zn), and 324.7 nm. The number seen in the spectrophotometer is stated as a metal value.

The water physicals and chemicals measured were color, temperature, pH and DO. In details, the color was observed visually, the temperature was measured by the thermometer, pH was measured using the pH meter, and dissolved oxygen (DO) was measured using DO meter.

The data of heavy metals concentration (Hg, Cd, Pb, Zn, and Cu) in water and sediment was tested by using ANOVA and analyzed descriptively. On the other hand, the statistical analysis was done using SPSS version 16.0 software.

RESULTS AND DISCUSSION

Metal types that are identified in the water flowed with waste. The results of the observations show that there are mercury (Hg), cadmium (Cd), lead (Pb), zinc (Zn), and copper (Cu) in all sampling points because the waste that passes through the water stream coming from household waste as well as agricultural and industrial waste containing organic and inorganic materials and heavy metals. Heavy metals which are mercury (Hg), cadmium (Cd), chromium (Cr), lead (Pb), copper (Cu) have high toxicity and will accumulate in the body of an organism (Murty et al. 2013). The heavy metal contamination such as Pb, As, Cd, Hg, Cr, Ni occurs in various water sources such as soil, surface, tap water etc. Some of those heavy metals are potentially toxic and transferred to the surrounding environment through different pathways (Sankhla et al., 2016)

The main sources of mercury are anthropogenic (Patra & Sharma, 2000) originating from the use of metals in industry, mining, batteries, and mercury lamps. Mercury (Hg) is one of the most toxic heavy metals compared to other heavy metals (Zhang et al., 2017) because it can turn into methyl mercury which is more toxic than other mercury types (Gautam, Sharma, Mahiya, & Chattopadhyaya, 2014). Mercury that accumulates in organisms will disturb the growth and development of the organism (the United States Environmental Protection Agency, 2010) and even able to cause death (Clarkson & Magos, 2006).

Cadmium (Cd) is one of the non-essential heavy metals which has a relatively small amount but can be increased in environments that are contaminated with industrial waste (Pacyna, 2010). Cadmium can be found in phosphate fertilizer as a source of pollutants in water (Jarup, 2014). The main route of Cd is the respiratory and digestive systems (Terry and Stone, 2002).

On the one hand, lead (Pb) is included in the non-technical metal which has toxic nature and is not useful for the organism; tend to be poisonous for the organism (Suhendrayatna, 2001; Yorulmazlar and Gul, 2003). Lead content in water is originated from chemical waste, printing industry waste, paint industry waste, and other industries which produce heavy metals (Sumah and Aunurohim, 2013). Lead (Pb) is a poison for metabolic and enzyme inhibitors (Gebrekidan M., Samuel, 2011) that can damage nervous connections, blood, and brain disorders, as well as hematologic damage (Mohod and Dhote, 2013).

Zinc (Zn) is an essential nutrient for humans and all living creatures. Zinc (Zn) is a cofactor for more than 300 enzymes and is found in all tissues. In humans, the highest concentration of zinc is found in bone, muscle, prostate, liver, and kidneys. A similar distribution has been found in animals that less than 10% of the total zinc in the body is in the blood (EPA, 2005) while the rest is in bone and muscle.

Copper ions usually react with inorganic and organic chemicals in solutions and suspensions which then will form complex to highly complex solutes and sediments containing many natural inorganic and organic water constituents (carbonates, phosphates, and organic substances) (EPA, 2016). Copper (Cu) at low concentration is a micronutrient for organisms and is essential for almost all plants and animals (Kapustka et al. 2004). It is a key atom in protein which acts as a carrier of oxygen (hemocyanin) from several invertebrates. However, Cu can be toxic to some aquatic life at high concentrations. Cu toxicity depends on the organism. In bacteria, fungi, and algae, excess copper interferes cell membrane integrity and causes subsequent leakage of cell content (Borkow and Gabbay 2005).

Metal concentration in water. The test results illustrate that the concentration of mercury (Hg) is ranged from 0.028±0.002 mg/L to 0.036±0.005 mg/L while lead (Pb) is in between 0.492±0.009 mg/L and 0.665±0.007 mg/L, cadmium (Cd) in between 0.142±0.011 mg/L and 0.161±0.004 mg/ L, zinc (Zn) in between 0.737±0.006 mg/L and 0.763±0.004 mg/L), and copper (Cu) in between 0.318±0.004 mg/L and 0.369±0.002 mg/L. The highest concentration of Zn compared to other metals and all metal concentrations (Hg, Pb, Cd, Zn, and Cu) was found in the S2 sample. The concentration of heavy metals at all sample points from highest to lowest consecutively is Zn, Pb, Cu, Cd, and Hg (Figure 1).



Figure 1 – The concentration of metals in water

The S2 sample is a sample taken right at the pollutant source, making it possible to contain the highest metal concentration. The distance of pollutant sources determines the concentration of Hg that is accumulated. The closer the distance, the higher the concentration (Musthofa et al., 2016). Some of the heavy metals that enter the water will be carried by the water stream whereas some will settle in the sediment.

Among other heavy metals, Zn has the highest concentration in the water stream because zinc (Zn) is an essential metal needed by organisms in metabolic processes. Zinc enters the body of the organism through food and water to be used in the process of biotransformation and bioaccumulation (Palar 2008). Meanwhile, mercury (Hg), cadmium (Cd), lead (Pb), and copper (Cu) are known to contain toxic materials (Duruibe, *et al* ., 2007) which have high toxicity and will accumulate in the body of the organism (Murty, *et al*, 2013).

The maximum threshold of concentration of all metals (Hg, Pb, Cd, Zn and Cu) is recommended by the Government Regulation of the Republic of Indonesia number 82 (2001) as follows Hg 0.002 mg/L, Pb 0.03 mg/L, Cd 0.01 mg/L, Zn 0.05 mg/L, and Cu 0.02 mg/L. From the test results, it is shown that the concentration of all heavy metals (Hg, Pb, Cd, Zn, Cu) along the water stream has exceeded the recommended maximum threshold.

Heavy metals concentration in sediments. The test results show that the concentration of mercury (Hg) in the sediment is ranged from 0.141±0.007 mg/L to 0.268±0.006 mg/L while lead (Pb) is in between 1.380±0.004 mg/L and 1.453±0.005 mg/L, cadmium (Cd) in between 0.559±0.002 mg/L and 0.570±0.003 mg/L, Zn in between 1.782±0.003 mg/L and 1.827±0.006 mg/L, and copper (Cu) in between 1.153±0.004 mg/L and 1.363±0.004 mg/L. The quality standard for heavy metal concentrations in sediments from the *International Association of Dredging Companies/the Central Dredging Association* (IACD/CEDA) (1997) is 85 mg/L - 1000 mg/L. Heavy metals in sediments are still below the target level so it is assumed not too dangerous for the environment.

The highest heavy metal concentration for Hg, Pb, Cd, Zn, and Cu occurs in sample S2 whereas the lowest concentration for Hg and Cu occurs in sample S3. On the other hand, the lowest concentration of Pb, Cd, and Zn is found on sample S1. In fact, Zn dominates the sediment while Hg is known to be the lowest (Figure 2).

It can be seen that the highest concentration of heavy metals (Hg, Pb, Cd, Zn, Cu) in all samples lies in the S2 sample (Figure 3). The metal concentration on sediment is reported to be higher than in the water. This happens because some of the heavy metals which enter the water stream are carried away by the flow and are accumulated in sediment. Heavy metals such as Pb, Cd, Cu, and Hg can interact with organic substances in the form of a solution which then will be sedimented causing a high concentration in the sediments (Begum et al., 2009b). This will continue to accumulate with the increasing waste in the water (Begum et al., 2009a). On the other hand, heavy metals in water tend to follow the flow of the water stream and will dilute when a new flow enters so that the concentration decreases.



Figure 2 – The concentration of metals in sediment



Figure 3 – The concentration of metals (Hg, Pb, Cd, Zn, Cu) in all sample

All heavy metals at the sampling points indicate a positive correlation between the concentration of heavy metals in water and sediments. In details, mercury (Hg) in water affects mercury in the sediment by 80.4% with the equation Y = 1.357 + 1.374X. Whereas, lead (Pb) in water affects the Pb in sediments by 81.7% with the equation Y = 0.181 + 0.143X. The concentration of Cd in water affects the concentration of Cd in sediments by 82.1% with the equation Y = 0.147 + 0.123X. Then, Zinc (Zn) in water affects Zn in sediments by 81.8% with the equation Y = 0.331 + 0.586X. Last but not least, Copper (Cu) in water affects Cu in the sediment by 98.2% with the equation Y = 0.605 + 1.094X.

The concentration of heavy metals in water will affect the concentration of heavy metals in the sediment. The increased concentration of heavy metals in sediments is caused by the high concentration of metals which entered the water. Metals that are hard to dissolve will go through a dilution process in the water, drop to the bottom of the water stream, and settle in the sediment (Facetti et al., 1998). Heavy metals in water become part of the water-sediment system that their distribution is controlled by dynamic equilibrium and physics-

chemical interactions influenced by pH, concentration, and type of compound (Singh, Mohan, Singh, & Malik, 2005) (Singh et al., 2005). The metal binding in sediment is caused by the electrochemical attraction between sediment particles and metal particles and the metal binding in organic particles (Meregalli, *et al.*, 2004).

Water quality. The physical and chemical components of the water observed include temperature, acidity (pH), and dissolved oxygen (DO). The measurement of environmental parameters is important because it influences the absorption rate of heavy metals in the water (Darmono, 2000). The results of the measurement imply that the temperature is ranged from 27.3 - 29.7°C while the pH is in between 5.67 - 6.18 and DO in between 4.51 mg/L – 5.37 mg/L. In this case, the water looks muddy.

The results of the water physical and chemical components measurement point out that the temperature is still within the normal range and the pH tends to be acidic to neutral. Besides that, it is known that the DO is still within the recommended maximum value range (RI Government Regulation number 81 of 2001) (0-6 mg/L). The color of the water is light brown to dark brown which indicates that the water stream is muddy because of the many organic particles contained in the water.

Temperature influences the solubility of oxygen in water; when the temperature increases, the oxygen solubility in water will be low (EPA, 2001). The amount of dissolved oxygen (DO) signifies the level of pollution; when the oxygen is high, the level of pollution will be low and vice versa (Mccaffrey, 1995). Dissolved oxygen will decrease if the water is polluted because there is a decomposition process of organic substance that requires oxygen (Ann, 2002).

CONCLUSION

It is concluded that the water stream in Desa Kejapanan, Pasuruan contains mercury (Hg), lead (Pb), cadmium (Cd), zinc (Zn), and copper (Cu) originating from domestic, agricultural, and industrial waste. The waters are indicated to have been contaminated by heavy metals (Hg, Pb, Cd, Zn, Cu) because the concentrations of these metals have exceeded the maximum threshold both in water and in sediments.

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